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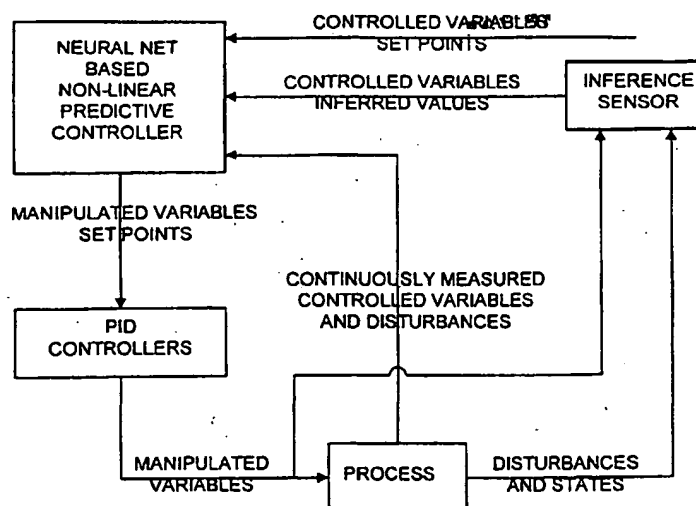
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(54) Title: **PROCESS FOR THE CONTROLLED PRODUCTION OF POLYETHYLENE AND ITS COPOLYMERS**



(57) Abstract: A process for producing high density polyethylene (HDPE) or linear low density polyethylene (LLDPE) in gas phase reactors, isolated or combined, using chromium or Ziegler-Natta catalysts, provided with on-line control of certain process variables as well as physical properties of the produced resin is described. Such process comprises the use of models for the inference of the physical properties and of the process variables that are not continuously measured as well as models which are relevant for the control of said properties and of the operating conditions of the process. The control of the process variables provides further the maximization of the production rate and of the catalyst productivity in the polymerization reaction.

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**PROCESS FOR THE CONTROLLED PRODUCTION OF  
POLYETHYLENE AND ITS COPOLYMERS  
FIELD OF THE INVENTION**

The present invention relates to a process for the controlled  
5 production of high density polyethylene (HDPE) or of linear low density  
polyethylene (LLDPE) in one or more gas phase reactors, in the presence of  
either chromium or Ziegler-Natta catalysts, said process possessing an on-  
line control of certain process variables as well as the control of certain  
physical properties of the final resin product. More specifically, such  
10 process comprises: i) the use of models for inferring the physical properties  
as well as the process variables which are not measured continuously and  
ii) models which are relevant for the control of said properties and  
operating conditions of the process under study. The control of the process  
variables provides further the maximization of the production rate as well  
15 as of the catalyst yield in the polymerization reaction.

**BACKGROUND INFORMATION**

The control of the process variables in petrochemical plants is  
normally manually effected by operators who periodically sample the  
product to be tested and act to maintain or to correct the operating  
20 conditions as to obtain a product having the desired characteristics. This  
involves delays related to required corrections, since the sampling and  
laboratory tests normally lag the on-line process, besides possible human  
errors. Further, the dynamics of petrochemical processes is generally slow.  
Thus, long periods of time may be required so that the effect of the  
25 adjustments performed on the process input variables become effective.  
Therefore, the slow dynamics is a feature which renders difficult the

control of the industrial unit since the worker may not know if there has been a sufficient time for the effects of the performed adjustments to be apparent or either if more time is required so that said effects are completely perceived.

5           On the other hand, techniques for the empirical as well as rigorous modeling are known, which may be used for obtaining process models. Such mathematical models are able to infer the value of certain process variables that are periodically measured from other process variables that are continuously measured. Besides, the mathematical models  
10 may also be used to predict the future behavior of process variables caused by modifications in the operating conditions of an industrial plant.

          The techniques of rigorous modeling are based on the physical principles, which describe the basic interactions between the process variables. As compared to the empirical techniques, the rigorous models are  
15 more difficult to construct and require a deep knowledge of the process. Besides, the complexity of the equations, which make up the rigorous model, may render it, at least in some cases, unsuitable for on-line implementation, this difficulty arising from the long time to resolve such equations, even making use of computers.

20           On the contrary, the techniques of empirical modeling do not require such a deep knowledge of the process being modeled and originate simpler mathematical models which may be quickly executed, being therefore suitable for being executed in real time. A disadvantage of the empirical models is that they cannot be used under operating conditions  
25 different from those used in their identification. Models obtained from linear and non-linear regressions as well as neural networks are among the

relevant empirical modeling techniques described in the literature.

Neural networks are networks of either neurons or elements which are interconnected in a unique way. Typically, the networks are made up of input neurons which receive signals or information from outside the network, output neurons which transmit signals or information to outside the network and at least one intermediate layer of neurons which receive and transmit the information to other neurons.

Besides the fact of a model being rigorous or empirical, it may also be characterized as static or dynamic. A model is said static when it yields as a result the steady state values of the process variables in view of the values assumed by the input variables of the system. On the other hand, the dynamic model presents, besides the information on the steady state of the system, information on the way by which the output variables move between two steady states.

A large number of references describe processes provided with control, for the production of polyolefins.

US Patent 3,636,326 teaches how to adjust the production rate of a polymerization reactor based on the catalyst yield calculated in real time. Thus, possible modifications in the catalyst yield may be automatically compensated by means of a feedback control loop. This kind of control may be practiced when the variables which affect the catalyst yield are not known or not measured. In this case the best to do is to automatically adjust the catalyst rate as soon as a variation in the catalyst yield is perceived. However, by using process models it is possible to preview that catalyst yield will undergo modifications due to certain changes in the operating conditions. Thus, it is possible to previously adjust

the catalyst rate so that the change in catalyst yield does not significantly affect the production rate of the reactor.

This and other advantages to be mentioned hereinbelow are features of the controlled process to be described and claimed in the present application.

US Patent 3,998,995 teaches a process in which the production rate and monomer concentration are controlled in a polymerization reactor able to restrict the concentration of the main monomer and the solids concentration at maximum values. In said process, the production rate is controlled by the flow rate of olefinic monomer while the concentration of monomer in the reaction zone is controlled through the adjustment of a diluent feed fed to the process. If the maximum limiting value for the monomer concentration is reached, monomer flow rate is then adjusted to reduce said concentration and no longer to control production. If the maximum limiting value for the solids concentration is reached, the flow rate of diluent is adjusted to reduce said concentration and no longer to control the monomer concentration. Whenever the constraints are violated the goal of controlling production rate and monomer concentration is abandoned in favor of the continuity of operation. The control system which is unable to deal with constraints will not be able to guarantee the safety and the operational continuity required for an industrial unit to accomplish its goals. In spite of the fact that the said US Patent presents a control procedure able to deal with the plant constraints, the system works only from the moment when the constraint has been effectively violated. A controller which could predict the future behavior of the process variables would be able to promote the required adjustments so that the constraint

would not be violated or would be just slightly violated. A process whose constraints are so controlled, would allow that the main control objectives would not be abandoned.

A method for the control of the intrinsic viscosity of a polymer which is being produced has been taught in US Patent 3,878,379. The technology unveiled in said US Patent, besides being specifically directed to the production of polyethylene terephthalate, relates to one single variable and does not contemplate the control of different polymer properties.

US Patent 4,469,853 provides a process for the production of polyolefins in the liquid phase having a few well defined properties. Such process describes the use of chromatography for the measurement of the concentrations of olefin monomers and hydrogen in the gaseous phase, which is formed on the top of the reactor. The control of the flow rates of ethylene and comonomers is adjusted so as to keep constant the ratio between the concentrations of said reagents so as to lead to a polyolefin having pre determined density. Further, said US Patent achieves the control of the ratios between the concentrations of hydrogen and main monomer by adjusting the flow rate of hydrogen feed to the reactor and thus leading to a polyolefin having a pre determined melt flow rate. Optionally, the partial pressure of ethylene in the reactor could be controlled through the adjustment of the flow rate of the feed of catalyst while the pressure could be controlled through the adjustment of the reactor flow rate of purge.

US Patent 5,098,967 teaches how to control the molecular weight of polypropylene and its copolymers produced in the liquid phase by measuring the heat released during the polymerization. The calculation of

the amount of polymerized monomer is effected on the basis of said measurement. This leads to the possibility to anticipate a controlled flow rate of hydrogen feed to the reactor so as to keep a pre determined ratio between reacted monomer and hydrogen feed so that it is possible to obtain  
5 a polymer of desired molecular weight.

US Patent 5,504,166 teaches how to control the melt flow rate and the comonomer content of a polymer produced in a horizontal stirred-bed reactor. Such properties are related to a set of operation variables and to parameters which are intrinsic to the process.

10 US Patent 5,844,054 teaches a process for the polymerization of olefins in gas phase reactors. The novelty of the process comprises the control of the production rate, of the density and of the melt flow rate of the resin by manipulating the flow rate of catalyst as well as the ratio between the flow rates of oxygen and monomer.

15 US Patent 5,282,261 teaches the use of values which are predicted by a neural network in real time to be used instead of a measuring instrument or a lab test as an input for a controller, so as to implement an inference and control system in a continuous process while using neural networks. However, said US Patent does not teach how to apply the  
20 described system to polymerization process nor teaches any control structure which might be particularly applied to the process for producing polyethylene and its copolymers.

Thus, none of the patents nor the publications of the state-of-the-art technique, either isolated or combined to each other, teach a process  
25 for producing HDPE or LLDPE in a single or combined gas phase reactors, using chromium or Ziegler-Natta catalysts in which process may be on-line

and simultaneously controlled the production rate, the catalyst yield, the superficial velocity and the gas composition within the reactor, among other process variables, together with various physical properties of the product, such as melt flow rate and density, such process being described and  
5 claimed in the present application.

### SUMMARY OF THE INVENTION

The present invention relates to a process for the controlled production of HDPE or LLDPE in a single or combined gas phase reactors, under polymerization conditions, in the presence of hydrogen, oxygen, inert  
10 diluents and using chromium or Ziegler-Natta catalysts, the process having an on-line control of certain process variables, as well as of some physical properties of the produced resin.

Such control is based on dynamic mathematical models which describe the effects of the manipulated variables and of the disturbances on  
15 the controlled variables as well as the variables whose values should be restricted to certain operation ranges. The use of such models renders possible to infer the properties of polyethylene in real time from the on-line measurement of other process variables. Besides providing an estimate of the quality of the polymer, models are used by an optimization algorithm  
20 which determines the best set of control actions to be taken so that the controlled variables can approach the set point for each of the variables without violating the constraints imposed to assure continuous and safe operation of the unit.

Thus, the present invention provides a process for the  
25 controlled production of polyethylene and its copolymers, the control being based on dynamic mathematical models used for the simultaneous and on-



line control of the melt flow rate 1 ( $MFR_1$ ), the ratio of melt flow rates ( $MFRR$ ) and the density of the resin produced from the chromium catalyst as well as the melt flow rate 2 ( $MFR_2$ ) and the density of the resin produced from Ziegler-Natta catalysts.

5           The present invention still provides a process endowed with an on-line control of the production rate, the catalyst productivity, the composition of the gas within the reactor, the superficial velocity of the fluidized bed system, the bubble temperature of the reactor recycle stream and the difference between the inlet and exit temperature of the water in the  
10   cooling system of the reactor.

          The present invention provides further the maximization of the production rate of polyethylene and the catalyst yield for the described process.

#### **BRIEF DESCRIPTION OF THE DRAWINGS**

15           FIGURE 1 is a representation of the categories in which are classed the process variables which make up a chemical process.

          FIGURE 2 is a representation of a control matrix.

          FIGURE 3 is a simplified representation of the plant for producing polyethylene according to the present invention.

20           FIGURE 4 is a schematic representation of the implementation of the process control described and claimed in the present invention.

          FIGURE 5 is a schematic representation of the implementation of the inference of the process variables.

          FIGURE 6 is a representation of the control matrix  
25   characteristic of the embodiment of the invention for the controlled production of polyethylene and its copolymers using a chromium catalyst.

FIGURE 7 is a representation of the control matrix characteristic of the embodiment of the invention for the controlled production of polyethylene and its copolymers using a Ziegler-Natta catalyst.

5           FIGURE 8 is a graph showing the frequency and intensity of the deviations between the production rate and its average value. The reduction in the variation of the production rate caused by the use of the process described and claimed in the present invention may be observed.

#### **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

10           In the present specification, the expression "on-line" means that the physical properties which are to be inferred are obtained in real time.

The expression "quality of the polymer" refers to the physical properties of the polymer such as melt flow rates  $MFR_1$  and  $MFR_2$ ,  $MFRR$ ,  
15   density etc., which affect the properties of the resin.

$MFR_1$  and  $MFR_2$  are the melt flow rates of the resin, measured according the ASTM Method D-1238. Measurements are made at 190°C using 21.6 g and 2.16 g weights, respectively.

$MFRR$  (melt flow rate ratio) is the ratio of melt flow rates of  
20   the resin, calculated as  $MFRR = MFR_1/MFR_2$

The density of the resin is measured according to ASTM Method D-1505.

The expression "catalyst productivity" is a synonym for catalyst yield and represents the mass of polymer produced per mass of  
25   catalyst used.

The expression SISO (Single Input Single Output) control strategy means that one single manipulated variable is adjusted to control a certain controlled variable.

The expression MIMO (Multiple Input Multiple Output) control strategy means that multiple manipulated variables are adjusted at the same time to control a set of controlled variables.

The term "feedback control" means the mechanism of feeding to a controller the difference between the desired value for a certain variable (set point) and the measured value of said variable.

The expression "gas phase reactor" means that in the reactor there are at least one gaseous phase containing monomer and at least one solid phase containing catalyst. The gas phase reactors useful for the practice of the invention are the vertical fluidized bed reactors or horizontal fluidized bed known in the art. The same way, other types, such as reactors provided with mechanical agitation, may be employed. A general idea of the kind of available technologies may be found in the chapter "Gas-phase Polymerization", Encyclopedia of Polymer Science and Engineering", second edition, vol. 7, p. 480-488, John Wiley & Sons, Ed.

A definition of Ziegler-Natta catalysts may be found in the chapter "Definitions, stereochemistry, experimental methods and commercial polymers", from the book by John Boor, Jr., "Ziegler-Natta Catalysts and Polymerizations", p. 32-35, Academic Press.

The chromium catalyst may be one of the well-known Phillips catalysts or either belong to another classification. A definition of the Phillips catalysts may be found in the chapter "Metal alkyl-free Catalysts" from the book by John Boor Jr, "Ziegler-Natta Catalysts and

Polymerizations", p.279- 324, Academic Press.

The present process is directed to the production of HDPE or LLDPE either as polyethylene homopolymer or copolymer. In case of copolymers, the comonomers may be chosen, besides ethylene, among the group of reactive olefins such as : propene; 1-butene; 2-methyl propene; 1-pentene; 1-hexene; 1-heptene; 1-octene; 1-decene; 3-methyl 1-butene; 4-methyl 1-pentene; cyclopentene, the comonomers being copolymerized isolated or in admixture.

The preferred catalyst for the production of HDPE is the chromium catalyst while the preferred catalyst for the production of LLDPE is the Ziegler-Natta catalyst. However, other catalysts may be employed as well.

According to FIGURE 1, in order to set a control strategy for polymerization reactors, it is convenient that the process variables are classified into: controlled variables (8), manipulated variables (1), constrained controlled variables or simply constraints (6) and disturbance variables or simply disturbances (7).

Controlled variables (CV) are those variables the value of which should be kept the closest possible to a desired value or set point. In a polymerization plant the controlled variables may be divided into two categories: polymer physical properties (4) and process variables which define operating conditions of the plant (5).

The physical properties (4) of the polymer such as melt flow rates  $MFR_1$  and  $MFR_2$ , density,  $MFRR$ , etc. establish the quality of the polymer. The process variables which define operating conditions of the plant (5) or simply operating conditions, involve temperatures, pressures,

flow rates, etc.

Constrained Controlled Variables CCVs are variables that, in spite of being controlled, do not need to have their value kept close to a set point. However, they should be controlled so as not to exceed certain limits.

5 Manipulated variables MVs are those which should be adjusted so that the controlled variables may be kept close to a set point or within certain limits. In order that a process variable is used as manipulated variable in a control strategy, the latter should somehow affect at least one controlled variable or a constrained variable. Examples of MVs are flow  
10 rates of monomer, catalyst and cooling water.

In general, operators who follow the operation of a chemical process know, at least qualitatively, the way a certain MV affects a certain CV. It is usual to have some limit in regard to the variation rate of MV since for certain variables it may not be safe to promote large adjustments  
15 in a short period of time.

Another reason for the existence of a limit or constraint in the variation rate is the inertness of the working instruments (valves, engines, etc.) which regulate the values of the MVs. Even if a valve is deemed to stay utterly open, there is a delay until the valve actuator moves from its  
20 present situation until the position where the opening of the valve is maximum. Thus, for a process control to be efficient and safe the variation rate constraint for the MVs should be taken into consideration.

Disturbance Variables DVs, are process variables which also affect the controlled variables but on which the plant operator cannot exert  
25 any action. Examples of disturbances are: impurities in the monomer stream which feeds the reactor and the catalyst quality. Disturbances

generally arise from known causes but it is not easy to eliminate them since they are originated by parameters which are extraneous to the process. Some disturbances may be measured, others may not. A control system may take into consideration the measured disturbances (2) by promoting an  
5 adjustment in the manipulated variables before the effect of the disturbances are apparent in the product quality.

The classification of a process variable into one of the above-mentioned categories is not always trivial. For example, the concentration of comonomer in the reactor may be an MV for the adjustment of the  
10 polymer density and at the same time be a CV in a concentration control loop which uses the comonomer flow rate as MV. In this case the MV for the control of the density is the desired value for the comonomer concentration. This kind of strategy is called cascade control. Besides, under the point of view of control, if a process input is not being used as  
15 MV then it may be considered as a DV. As indicated above, the classification of the variable as CV, CCV, MV and DV depends on the control loop being considered. Further, there is the fact that the same variable may be controlled and at the same time submitted to constraints. In the present specification the expression CV will be used for the variables  
20 the value of which should be kept close to a desired value (that is, the set point) even if said variable should be submitted to constraints.

The set of CVs, CCVs, MVs and DVs will make up the control matrix of the process which is one of the ways used to represent multivariable control systems. FIGURE 2 shows an example of control  
25 matrix. In this matrix the columns represent the CVs and CCVs and the lines represent the MVs and DVs. The assigned positions within the control

matrix indicate that a certain MV or DV is used in the modeling of a certain CV or CCV. It is the right choice of said variables which determines the scope of the control system as well as the potential benefits from its use in a controlled process.

5           Once the control matrix is defined, mathematical models of the process should be constructed for each CV and CCV, so that the model inputs should be the MVs and the DVs, while the outputs should be the CVs and the CCVs. The function of the models used for process control is precisely the storage of the cause-effect relationships between the  
10   CVs/CCVs and the MVs/DVs as mathematical equations.

          The physical properties of a polymer and the economical factors involved in the process, such as production rate and the catalyst yield, are a function of the raw materials used to make it, as well as of the operating conditions used in the polymerization reaction. Thus, once the  
15   raw materials to be used in the polymerization are determined, it is of utmost importance that the process inputs are manipulated so that the physical properties are controlled while at the same time high production rates and high catalyst yield are sought.

          Most of the physical properties which define the product  
20   quality are not measured on-line. In many cases there are no analyzers which could be directly connected to the process and there are cases where in spite of the fact that the technology for the measurement of such variables is available, the cost of the sensors may render its use impractical. This makes that tests are effected in the lab, with a sampling interval of a  
25   few hours. This way, during the period between one test and the following one, there is no indication whatsoever of the quality of the product, this

causing a gap between the possible alterations in the variables and the implementation of whatever corrections which might be deemed necessary. In order to overcome the problem of scarcity of measurements of the polymer quality, process modeling techniques may be used, those allowing  
5 to continuously infer the properties which are not continuously available.

A further relevant aspect related to the process control is that, in general, economical considerations cause that the process should be operated under operating conditions close to the plant limits. Thus, the flow rate of catalyst could be manipulated to control the reactor production rate;  
10 however, whenever the polymerization heat overcomes the limit of the heat exchange capacity of the reactor, the objective of a further increase in production should be restricted in favor of the thermal stability.

This example illustrates the importance of considering the CCVs in a multivariable control strategy since these are the variables which  
15 will determine the limits within which it is possible to safely operate the process. Besides, it is near these limits that the best result is obtained in economical terms, which may be visualized through the increased production rate as well as higher catalyst yield.

Polymerization processes show a high interaction degree  
20 among the various process variables, which renders difficult the task to pursue several control goals at the same time. One interaction example is the relationship existing among the control of density,  $MFR_1$  and the production rate in the process for producing polyethylene. Adjustments in the comonomer flow rate aiming at controlling the polymer density cause  
25 modifications in the production rate and the melt flow rate. In order to keep the specified melt flow rate, other MVs should be adjusted, which in turn



may intervene in the control of density and production rate. Thus, were the adjustments made independently, there would be a great possibility that a reduction or increase in the overall effect of the control actions on certain CVs could occur. In order to avoid such situation, the process adjustments  
5 should be performed considering the effect of all input variables (MVs and DVs) on the output variables (CVs and CCVs), as established in the control matrix so as to correct the value of the desired variable, without altering the value of the remaining variables.

Although it is possible to control the plant using static process  
10 models, this is not the most efficient alternative, since the adjustments should be performed slowly in view of the fact that the model does not contain information on the rate at which the process responds to said adjustments. On the contrary, a dynamic model-based system is able to perceive if the CVs are deviating from the desired operation condition even  
15 during transition periods. This is a further advantage of the controlled process of the present invention since in an industrial plant for producing polyethylene, transitions are frequent in order to make resins of varied specifications. The making of such transitions using the controlled process of the invention not only lowers the duration of the transition from one  
20 operating condition to the following one, but also minimizes the yield of off-specification resin.

Therefore, in order to obtain significant benefits, the control of such processes should be multivariable (MIMO), that is, it should comprise various CVs and various MVs. Besides, the process control should take into  
25 consideration the physical limits of the plant under study (CCVs and variation rates), possible DVs which may be measured as well as the

process dynamics.

Once the mathematical models have been adjusted to represent the relationships between the process variables, this model may be used for inference as well as for control.

5           In case it is used for inference, the model is designed to provide an estimation of the variables and physical properties which are not continuously available such as those which are measured through lab tests or through instruments in which data collection and measurement processes involve longer periods of time.

10           And in case it is used for control, the model is designed to predict the future behavior of the CVs and CCVs in case certain adjustments are made in the MVs and in case variations in the DVs might occur. Thus the controller may determine the best set of actions to be taken so that the controlled variables attain the desired values without violating  
15 the constraints.

          In the process which is the object of the present specification the modeling technique comprises preferably the use of neural networks as process models. Optionally, other empirical and/or rigorous modeling techniques may be used alone, combined among themselves or either  
20 combined to neural networks models for creating process models.

          FIGURE 3 presents the main streams and unit operations which make up a polyethylene plant in gas phase reactors operating in a fluidized bed. For the sake of illustration a vertical fluidized bed reactor is represented. The polyethylene production rate is controlled mainly through  
25 the injection of catalyst (stream 2). Monomer (stream 1) and, in case of the production of copolymer, comonomer(s) (stream 9) is fed to the system,

together with other constituents (stream 10) through the recycle stream of the reaction system (stream 8). The additional constituents comprise: hydrogen, oxygen, carbon dioxide besides inert diluents. Examples of inert diluents are: nitrogen, argon and up to 6 Carbon atoms alkanes, isolated or  
5 admixed in any amount. The gas composition within the reactor may be controlled by adjusting the flow rates of the various streams which are fed to the system. The flow rate of stream 8 should be such that the superficial velocity of the gas within the reactor is strong enough to support the bed without collapse and without entrainment of particles through the top of the  
10 reactor.

The non-reacted gas, excess monomer(s) and inert constituents when traversing the reactor keep the particles forming the bed in a fluidized state and remove the heat generated by the reaction. Said gas exits at the top of the reactor (stream 3) and adds to the monomer and constituents  
15 recovery stream (stream 6) so as to form stream 7. Such stream is compressed in compressor CP and cooled in heat exchanger HE. The flow rate of cooling water in such exchanger is manipulated so as to keep the reactor temperature stable. As polymerization proceeds, the product is continuously discharged from the reactor (stream 4). The rate of product  
20 withdrawal may be manipulated so as to keep the level of the fluidized bed stable. With the polymer, a certain amount of gas leaves the reactor with stream 4. In view of the fact that such diluents do not react, and in order to avoid the accumulation of those inert constituents in the system, there is a purge stream (stream 11) which may be used, together with the feed flow  
25 rates, to control the amount of inert constituents present in the system. Cyclone C separates the gas present in stream 4 and sends it back to the

reactor (stream 6) so that the monomer may be re-used. The polymer is directed (stream 5) to the degassing, deactivation and extrusion steps, which are not represented. Sometimes a portion of stream 8 enters the reactor partially in the liquid state. This way a portion of the reaction heat is removed as latent heat through the vaporization of the liquid present in the stream, leading to higher production rates.

When the plant is operated in the above-described manner, it is relevant to control the temperature of the bubble point of said stream, since if this value is much higher than the reactor temperature, because of an excessively high concentration of heavier constituents, there may be not enough heat to completely vaporize the liquid which enters the reactor, this causing operating drawbacks.

FIGURE 3 illustrates one possible configuration of the present process, which may be used either for producing HDPE or LLDPE. However, the present invention is not limited to the said embodiment, and may be applied to other kinds of gas phase reactors as well as to other configurations containing a higher number of reactors.

Among the main objectives of a plant for producing polyethylene and its copolymers are the preparation of polymers having physical properties within certain specifications and the optimization of economical goals such as a specific catalyst consumption and the production rate of the plant. That is, it is desired to minimize the consumption of catalyst per ton of produced polymer, this leading to increased catalyst productivity and to a reduction in the amount of catalyst residue in the product, as well as to the maximization of the amount of polymer produced per hour.

Conventionally, in such an industrial plant, the reactors are operated according to predetermined recipes and the lab tests show if there is the need to adjust the operating conditions so that the product quality is kept within the specified limits. The lab tests involve measurements of MFR<sub>1</sub>, MFR<sub>2</sub> and density. From the values of MFR<sub>1</sub> and MFR<sub>2</sub> it is possible to calculate MFRR, which serves as an indication of the molecular weight distribution of the resin.

According to the process of the invention, which controls the physical properties listed above, there is comprised a plant for producing polyethylene in the gas phase, a Digital Control System (DCS), a process computer connected to the DCS, an information system for the storage of historical process data and a system into which lab test values are fed.

FIGURE 4 shows, according to the inventive process, how the control is implemented. The controlled variables may be continuously available as in the case of the production rate or may be periodically measured as in the case of lab tests or chromatograph measurements. In case the CVs are periodically measured and/or present significant delays relative to the measurement period, the values of said variables are inferred in real time and those are the values which are controlled in the inventive process.

Whenever it is required to infer a property or a process variable one should use the procedure illustrated in FIGURE 5. The inputs of the mathematical models are measured in real time with the aid of the metering instruments connected to the

DCS. The DCS conveys the obtained values to the software installed in the process computer in charge of the calculations of the

mathematical model. The software that makes the calculations of the mathematical model also makes any pre-processing in the input signals as required. The output of the mathematical model is sent back to the DCS and also to the software which keeps the process historical data base so that  
5 those data can be used later on.

The inference of the desired property is obtained by multiplying the output of the mathematical model by a factor, called "ratio" in the present specification, which is calculated based on lab tests. The "ratio" is designed to correct possible mistakes in the estimation effected by  
10 the mathematical model when compared to the result of the lab test. This factor is calculated as the ratio between the value as supplied by the lab test and the value as estimated by the mathematical model at the moment when the tested sample was effectively collected. In order to avoid sudden variations in the inferred value, which is not good to the controller who  
15 receives such signals, the value should be filtered before the multiplication.

The "ratio" is calculated with the aid of the same program in charge of the process historical data base since, as mentioned hereinbefore, said calculation considers the value which is estimated by the mathematical model at the moment the tested sample was collected, such information  
20 being available only from the historical data base.

To correct the output of the mathematical model, a variation of the preferred embodiment comprises the use of another factor, hereinafter called "bias". Said factor is different from the ratio factor in that it should be added to the output value of the mathematical model. The bias is  
25 calculated as the difference between the value supplied by the test and the value estimated by the mathematical model at the moment when the tested

sample was effectively collected. The bias should also be filtered before its addition to the output of the mathematical model.

Once the values of the variables which make up the control matrix have been determined with the aid of instruments which are directly  
5 connected to the plant equipment, through lab tests or through calculations effected on the values of other measured variables, a problem of the non-linear optimization for determining the values that the MVs should take under a stationary regimen (set points of the MVs) is solved.

The solution of the optimization problem determines still the  
10 sequence of adjustments, which are to be effected throughout a pre-determined period of time, so that the MVs attain the desired calculated values when considering the dynamics and the importance of each one of the CVs, the variation rate and the admissible limits for each of the MVs so that the CVs are kept close to the desired values, without violating the  
15 constraints (CCVs and variation rate). PID controllers receive the set points from the MVs calculated by the predictive controller and are in charge of keeping such variables close to these values.

The maximization of the production rate may be effected by choosing an arbitrarily high set point for the production rate. Said CV is  
20 limited by the heat exchange capacity of the system, where the control is effected by the opening of the temperature adjustment valve, which serves as a constraint to the control system. The control system will increase the flow rate of catalyst until the opening of the temperature control valve reaches a safe limit for the operation of the unit. Were this constraint not  
25 followed, the flow rate of catalyst could increase excessively causing that the temperature control valve stayed completely open, with the

consequence that the temperature would be out of control and the unit would have to be shut down.

In spite of the fact that HDPE and LLDPE may be produced in the same unit, the production of each of these polymers has its own peculiarities. They are prepared in the presence of different catalysts, the HDPE being generally but not exclusively produced with a chromium catalyst while the LLDPE is generally but not exclusively produced with a Ziegler-Natta catalyst. Besides the catalyst type, the production of these polymers differs regarding to the operating conditions and also in regard to the procedures used in the operation of the plant. Due to such differences, this application describes and claims two embodiments or modes of the process for the controlled production of polyethylene, each of said modes using a specific control matrix according to the catalyst type used (chromium or Ziegler-Natta).

The process variables and the physical properties of the polyethylene, which may be controlled with the aid of the inventive process are:

- The MFR<sub>1</sub> of the resin;
- The density of the resin;
- The MFRR of the resin (for the chromium catalyst);
- The production rate;
- The catalyst productivity;
- The opening of the temperature control valve;
- The ethylene partial pressure;
- The superficial velocity of the gas in the fluidized bed;



- The ratio between the concentrations of hydrogen and ethylene;
- The ratio(s) between the concentrations of monomer(s) and ethylene;
- The bubble point of the recycle stream (for the Ziegler-Natta catalyst).

Thus, in one mode of the invention, the process for the controlled production of polyethylene in gas phase reactor(s), isolated or combined with each other, under polymerization conditions, in the presence of hydrogen, oxygen, inert diluents and a chromium catalyst should comprise:

a) in the laboratory, periodically testing the MFR<sub>1</sub>, MFRR and the density of the resin;

b) establishing the desired values or limits for the set of CVs and CCVs which encompasses production rate, MFR<sub>1</sub>, MFRR and the density of the resin, catalyst productivity, opening of the temperature control valve, monomer partial pressure and superficial velocity of the gas in the reactor;

c) determining, in real time, the value of the variables cited in item b), the properties of the resin having their value continuously inferred with the aid of mathematical models;

d) establishing limits for the set of MVs which encompasses flow rate of catalyst, ratio between the concentrations of one or more comonomers and ethylene, ratio between the flow rates of one or more comonomers and ethylene, temperature of the fluidized bed, ratio between the flow rates of oxygen and ethylene, ratio between the flow rates of hydrogen and ethylene, flow rate of the recycle stream, flow rate of inert diluent(s) and opening of the purge valve;

e) establishing limits for the variation rate of the variables of item d);

f) using mathematical models of the process, simultaneously calculating the values which should be assumed by the variables cited in item d) under a stationary regimen, considering that the level of the fluidized bed as well as the reactor pressure will be kept at constant values, so that the desired values set forth in b) will be reached without violating the limits established in items b) and d);

g) determining the sequence of adjustments, to be effected during a pre determined period of time, so that the variables cited in d) reach the desired values calculated in item f) considering the dynamics and the importance of each of the variables cited in b) as well as the constraints established in e);

h) adjusting the flow rates of catalyst, comonomer(s), cooling water, oxygen, hydrogen, inert diluent(s), the flow rate of the recycle stream and the opening of the purge valve so as to satisfy the previous items; and

i) based on the lab tests, periodically correcting the value which is inferred for the properties of the resin.

Presented below, the input variables for each of the models comprised for the production of polyethylene based on the chromium catalyst according to the present invention. The relationships described below are summarized in FIGURE 6. The names of the process variables which are shown in said Figure are described in TABLE 1.

The preferred comonomer, the flow rate of which is used as MV, is 1-butene, but optionally other comonomers may be used as well.

**Melt Flow Rate (MFR<sub>1</sub>)**

This property is a controlled variable, generally related to the molecular weight and to the polymer composition. In the state-of-the-art technique such property is assessed through periodical collections, this causing a time gap between the instant of the measurement and what actually occurs in the plant.

In case the MFR<sub>1</sub> model of the resin is designed to the inference or to the control of said property, the relevant parameters are:

- temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the hydrogen and ethylene concentrations;
- ratio between the comonomer(s) and ethylene concentrations.

**Ratio of Melt Flow Rates (MFRR)**

In case the MFRR model of the resin is designed to the inference or to the control of said property, the relevant parameters are:

- temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the hydrogen and ethylene concentrations;
- ratio between the comonomer(s) and ethylene concentrations.

**Density**

In case the model of density of the resin is designed to the inference or to the control of said property, the relevant parameters are:

- temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the comonomer(s) and ethylene concentrations.

### **Production Rate, Opening of the Temperature Control Valve and Catalyst Productivity**

These variables are of utmost importance for the process economics. The lower variance of said variables resulting from a more advanced control method such as that described and claimed in the present invention causes that the average values of said variables are higher than those obtained from the conventional control systems, taking into consideration the heat transfer limits of the reactors, which are represented by the opening of the temperature control valve.

The opening of the temperature control valve is one of the constraints which should be taken into consideration in order not to miss the reaction control, since if said valve is completely open there is no possibility to increase the flow rate of the cooling water to the heat exchanger and the temperature goes off, this causing that the unit should be shut down.

The control models for the production rate, opening of the temperature control valve and catalyst productivity use the following input variables to predict the future behavior of the said variables:

- flow rate of catalyst;
- ratio between the flow rates of comonomer(s) and ethylene;
- temperature of the fluidized bed;
- ratio between the flow rates of hydrogen and ethylene;
- flow rate of inert diluent(s);
- opening of the purge valve;
- level of the fluidized bed.

**Ethylene Partial Pressure**

The control models for ethylene partial pressure use the following input variables to predict the future behavior of the said variable:

- ratio between the flow rates of comonomer(s) and ethylene;
- 5     · temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the flow rates of hydrogen and ethylene;
- reactor pressure;
- opening of the purge valve;
- 10    · level of the fluidized bed.

**Ratio between the flow rates of hydrogen and ethylene in the reactor**

The control model for the ratio between the concentrations of hydrogen and ethylene in the reactor uses the following input variables to predict the future behavior of the said variable:

- 15     · ratio between the flow rates of comonomer(s) and ethylene;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the flow rates of hydrogen and ethylene;
- flow rate of inert diluent(s);
- opening of the purge valve.

**20    Ratio between the concentrations of comonomer(s) and ethylene in the reactor**

The control model(s) for the ratio between the concentrations of comonomer(s) and ethylene in the reactor use the following input variables to predict the future behavior of the said variable(s):

- 25     · ratio between the flow rates of comonomer(s) and ethylene;

- temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the flow rates of hydrogen and ethylene;
- flow rate of inert diluent(s);
- 5       · opening of the purge valve.

### Superficial Velocity

The control model for the superficial velocity in the reactor uses the following input variable to predict the future behavior of the said variable:

- 10       · ratio between the flow rates of comonomer(s) and ethylene;
- temperature of the fluidized bed;
- ratio between the flow rates of oxygen and ethylene;
- ratio between the flow rates of hydrogen and ethylene;
- flow rate of the recycle stream;
- 15       · flow rate of inert diluent(s);
- opening of the purge valve.

And according to another preferred mode of the invention, the process for the controlled production of polyethylene and its copolymers in gas phase reactor(s), isolated or combined with each other, under  
20 polymerization conditions, in the presence of hydrogen, oxygen, inert diluents and a Ziegler-Natta catalyst should comprise:

(a) In the laboratory, periodically testing the  $MFR_2$  and density of the resin,

(b) Establishing desired values or limits for the set of CVs and  
25 CCVs which comprise production rate, the resin  $MFR_2$  and density, catalyst productivity, opening of the temperature control valve, monomer partial

pressure, superficial velocity of the gas in the reactor, bubble point of the recycle stream and difference between the inlet and exit temperature of the water in the cooling system of the reactor;

- (c) Determining in real time the value of the variables of item  
5 (b), the resin properties having their value continuously inferred with the aid of mathematical models;

- (d) Establishing limits for the set of MVs which comprises  
flow rate of catalyst, ratio between the concentrations of one or more  
comonomers and ethylene, ratio between the flow rates of one or more  
10 comonomers and ethylene, temperature of the fluidized bed, ratio between  
the flow rates of oxygen and ethylene, ratio between the concentrations of  
hydrogen and ethylene, ratio between the flow rates of hydrogen and  
ethylene, flow rate of the recycle stream, flow rate of inert diluent(s) and  
opening of the purge valve; reactor pressure, level of the fluidized bed,  
15 density of the gas in the recycle stream and superficial velocity;

- (e) Establishing limits for the variation rate of the variables  
cited in item (d);

- (f) By using mathematical models of the process,  
simultaneously calculating the values that the variables cited in item (d)  
20 should take in a stationary condition, considering that the level of the  
fluidized bed and the reactor pressure will remain constant, so that the  
desired values established in (b) will be reached without violating the limits  
established in items (b) and (d);

- (g) Determining the sequence of adjustments to be made along  
25 a predetermined period of time, so that the variables cited in item (s) reach  
the desired values calculated in item (f) considering the dynamics and the

relevance of each of the variables cited in (b) and the constraints established in (e);

(h) Adjusting the flow rates of catalyst, comonomer(s), cooling water, oxygen, hydrogen, inert diluent(s), the flow rate of the recycle stream and the opening of the purge valve so as to satisfy the previous items;

(i) based on the laboratory tests, periodically correcting the value inferred for the resin properties

The input variables for each of the models comprised by the process for producing polyethylene using a Ziegler-Natta catalyst according to the present invention are presented below. The relationships described hereinafter are summarized in FIGURE 7. The name of the process variables which appear in such Figure are described in Table 1.

The comonomer(s) preferably used, the flow rates of which are used as MV are 1-butene and optionally 1-hexene, but other comonomers may be used as well, according to the resin to be produced.

#### **Melt Flow Rate 2 (MFR<sub>2</sub>)**

In case that the MFR<sub>2</sub> model of the resin is designed for the inference or for the control of said property, the relevant parameters are:

- Temperature of the fluidized bed;
- Ratio between the concentrations of hydrogen and ethylene;
- Ratio between the concentration(s) of comonomer(s) and ethylene.

#### **Density**

In case the model of the density of the resin is designed for the inference or for the control of said property, the relevant parameters are:



- Temperature of the fluidized bed;
- Ratio between the concentrations of hydrogen and ethylene;
- Ratio between the concentration(s) of comonomer(s) and ethylene.

5    **Production rate, opening of the temperature control valve, difference between the inlet and exit temperatures of the water in the cooling system of the reactor and ethylene partial pressure.**

Besides the opening of the valve there is another factor which should be considered when the process stability is sought, this being the  
10    difference between the inlet and exit temperatures of the water in the reactor cooling system. The water exits the heat exchanger at a temperature higher than the inlet temperature and is directed to a cooling tower so that it may return to the heat exchanger and take back the reaction heat from the system. Said cooling tower may receive hot water from various heat  
15    exchangers. If the water gets to the cooling tower at a very high temperature the tower does not succeed in cooling the water as required and said water returns to the heat exchange system of the reactor at a temperature which is still too high. If the temperature of the water which is directed to the heat exchanger is too close to the temperature at which the  
20    water exits the heat exchanger, the temperature gets out of control since the water which reaches the heat exchanger loses the capacity of withdrawing heat. Generally in this case the temperature control valve eventually is completely open in trying to withdraw more heat, however this is useless since there is no point in opening the temperature valve if the water which  
25    enters the heat exchanger is still too hot.

The control models for production rate, opening of the temperature control valve, difference between the inlet and outlet temperatures of the water in the cooling system of the reactor and ethylene partial pressure use the following input variables to predict the future behavior of said variables:

- Flow rate of catalyst;
- Temperature of the fluidized bed;
- Ratio between the flow rates of comonomer(s) and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Reactor Pressure
- Opening of the purge valve;
- Level of the fluidized bed.

#### **Catalyst productivity**

The control models for the catalyst productivity use the following input variables to predict the future behavior of said variable:

- Temperature of the fluidized bed.
  - Ratio between the flow rates of comonomer(s) and ethylene;
  - Ethylene partial pressure.
- Ratio between the concentrations of hydrogen and ethylene in the reactor**

The control model for the ratio between the concentrations of hydrogen and ethylene in the reactor uses the following input variables to predict the future behavior of said variable:

- Ratio between the flow rates of comonomer(s) and ethylene;

- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Opening of the purge valve;
- Flow rate of catalyst;
- 5       · Temperature of the fluidized bed;
- Ethylene partial pressure;
- Level of the fluidized bed.

**Ratio between the concentration(s) of comonomer(s) and ethylene in the reactor**

- 10       The control model for the ratio between the concentrations of comonomer(s) and ethylene in the reactor uses the following input variables to predict the future behavior of said variable:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- 15       · Flow rate of inert diluent(s);
- Opening of the purge valve;
- Flow rate of catalyst;
- Ethylene partial pressure;
- Level of the fluidized bed.

20       **Bubble point of the recycle stream**

      The control model for the bubble point of the recycle stream in the reactor uses the following input variable to predict the future behavior of said variable:

- Ratio between the flow rates of comonomer(s) and ethylene.

25       **Superficial Velocity**

The control model for the superficial velocity in the reactor uses the following input variables to predict the future behavior of said variable:

- Flow rate of the recycle stream;
- Density of the gas in the recycle stream.

Having improved the control of the production rate in the reactor a reduction in the standard deviation of said variable was observed. As a consequence of the better stability, the average production rate of the unit was increased, as demonstrated in the Example below.

TABLE 1 below lists the variables involved in the control matrices represented in FIGURE 6 and FIGURE 7.

TABLE 1

VARIABLE	DESCRIPTION
PRODUCTION	Production rate
MFR <sub>1</sub>	Melt Flow Rate 1 of the polymer
MFR <sub>2</sub>	Melt Flow Rate 2 of the polymer
MFR <sub>R</sub>	Ratio of Melt Flow Rates of the polymer
Density	Density of the polymer
PRODUCTIVITY	Catalyst Productivity (ton PE/kg cat)
TV	Opening of the temperature control valve
PP	Monomer Partial Pressure
SUP VEL	Superficial Velocity
CAT	Flow rate of catalyst
DELTA T	Difference between the inlet and exit temperature of the water in the reactor cooling system
C <sub>2</sub> /C <sub>1</sub>	Ratio between the flow rates of comonomer(s) and ethylene
TEMP	Temperature of the fluidized bed
RO	Ratio between the flow rates of oxygen and ethylene
H <sub>2</sub> /C <sub>2</sub>	Ratio between the flow rates of hydrogen and ethylene
RECYCLE	Flow rate of the recycle stream
IN	Flow rate of inert diluent (s)
PRESSURE	Reactor pressure
BO	Opening of the purge valve
LEVEL	Level of the fluidized bed
[H <sub>2</sub> ]/[C <sub>2</sub> ]	Ratio between the concentrations of hydrogen and ethylene
[C <sub>2</sub> ]/[C <sub>1</sub> ]	Ratio between the concentrations of comonomer(s) and ethylene
BP	Bubble point of the recycle stream
GAS DENS	Density of gas in the recycle stream

**EXAMPLE**

One of the main benefits consequent to the use of the advanced control method as described in the present invention refers to the lower  
5 variability in the product quality and in process variables such as production rate. The standard deviation of the production rate for the reactor under study was reduced from 0.62 ton/h to 0.27 ton/h, this meaning a 56% reduction. This is illustrated in FIGURE 8.

The higher shift between the production rate and its average,  
10 which in the conventional process is around 22%, was reduced to around 10%. This result was obtained by comparing the production rate of the unit during 7 days continuous operation with and without the control method of the invention.

Besides the reduction in the variability of the production rate  
15 under normal conditions, it may be seen that if an out-of-control situation for the reactor temperature occurs, the normal production rate is quicker and more efficiently recovered.

The sum of these factors make possible that the average of the production rate be shifted to higher values. For the reactor under study the  
20 increase in said variable reached 19.2%.

### CLAIMS

1. A process for the controlled production of polyethylene and its copolymers in gas phase reactor(s), isolated or combined, under polymerization conditions, in the presence of hydrogen, oxygen, inert  
5 diluent(s) and chromium catalyst, wherein said process comprises the steps of:

a) In the laboratory, periodically testing the  $MFR_1$ ,  $MFRR$  and the density of the resin;

b) establishing the desired values or limits for the set of CVs  
10 and CCVs which comprises production rate,  $MFR_1$ ,  $MFRR$  and the density of the resin, catalyst productivity, opening of the temperature control valve, monomer partial pressure and superficial velocity of the gas in the reactor;

c) determining, in real time, the value of the variables cited in item b), the properties of the resin having their value continuously inferred  
15 with the aid of mathematical models;

d) establishing limits for the set of MVs which comprises flow rate of catalyst, ratio between the concentrations of one or more comonomers and ethylene, ratio between the flow rates of one or more comonomers and ethylene, temperature of the fluidized bed, ratio between  
20 the flow rates of oxygen and ethylene, ratio between the flow rates of hydrogen and ethylene, flow rate of the recycle stream, flow rate of inert diluent(s) and opening of the purge valve;

e) establishing limits for the variation rate of the variables of item d);

25 f) using process mathematical models, simultaneously calculating the values which should be assumed under a stationary regimen

by the variables cited in item d), considering that the level of the fluidized bed as well as the reactor pressure will be kept at constant values, so that the desired values set forth in b) will be reached without violating the limits established in items b) and d);

5           g) determining the sequence of adjustments, to be effected during a pre determined period of time, so that the variables cited in d) reach the desired values calculated in item f) considering the dynamics and the importance of each of the variables cited in b) as well as the constraints established in e);

10           h) adjusting the flow rates of catalyst, comonomer(s), cooling water, oxygen, hydrogen, inert diluent(s), the flow rate of the recycle stream and the opening of the purge valve so as to satisfy the previous items; and

          i) based on the lab tests, periodically correcting the value  
15   which is inferred for the properties of the resin.

2. A process according to claim 1, wherein the gas phase reactors are vertical or horizontal fluidized bed or vertical or horizontal stirred-bed reactors.

3. A process according to claim 1, wherein the mathematical  
20   models may be empirical, rigorous or a combination of empirical and rigorous models.

4. A process according to claim 1, wherein the empirical mathematical models are neural networks.

5. A process according to claim 1, wherein ethylene is  
25   polymerized alone or combined with one or more olefin comonomers.

6. A process according to claim 5, wherein the olefin comonomers are propene; 1-butene; 2-methyl propene; 1-pentene; 1-hexene; 1-octene; 1-decene; 3-methyl 1-butene; 4-methyl 1-pentene; cyclopentene.

5           7. A process according to claim 1, wherein the inert diluent(s) are nitrogen; argon and alkanes having up to 6 carbon atoms, isolated or in admixture with each other in any amount.

8. A process according to claim 1, wherein the physical property of the resin that is controlled is the melt flow rate.

10           9. A process according to claim 8, wherein the model for the control of the melt flow rate of the resin uses the following input variables:

- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- 15           · Ratio between the flow rates of comonomer(s) and ethylene.

10. A process according to claim 1, wherein the physical property of the resin that is controlled is the ratio of melt flow rates.

11. A process according to claim 10, wherein the model for the control of the melt flow rate of the resin uses the following input variables:

- 20           · Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
  - Ratio between the flow rates of hydrogen and ethylene
  - Ratio between the flow rates of comonomer(s) and ethylene.

12. A process according to claim 1, wherein the physical  
25   property of the resin that is controlled is the density.



13. A process according to claim 12, wherein the model for the control of the density of the resin uses the following input variables:

- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- 5     · Ratio between the flow rates of comonomer(s) and ethylene.

14. A process according to claim 14, wherein the variable that is controlled is the production rate.

15. A process according to claim 14, wherein the model for the control of the production rate uses the following input variables:

- 10     · Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- 15     · Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

16. A process according to claim 1, wherein the variable that is controlled is the opening of the temperature control valve.

20     17. A process according to claim 16, wherein the model for the control of the opening of the temperature control valve uses the following input variables:

- Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- 25     · Temperature of the fluidized bed;

- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

5

18. A process according to claim 1, wherein the variable that is controlled is the catalyst productivity.

19. A process according to claim 18, wherein the model for the control of the catalyst productivity uses the following input variables:

10

- Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

15

20. A process according to claim 1, wherein the variable that is controlled is the partial pressure of ethylene in the reactor.

20

21. A process according to claim 20, wherein the model for the control of the partial pressure of ethylene in the reactor uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene

25

- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

22. A process according to claim 1, wherein the variable that is controlled is the ratio between the concentrations of hydrogen and ethylene in the reactor.

23. A process according to claim 22, wherein the model for the control of the ratio between the concentrations of hydrogen and ethylene in the reactor uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Opening of the purge valve.

24. A process according to claim 1, wherein the variable that is controlled is the ratio between the concentrations of comonomer(s) and ethylene in the reactor.

25. A process according to claim 24, wherein the model for the control of the ratio between the concentrations of comonomer(s) and ethylene in the reactor uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene

- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Opening of the purge valve.

26. A process according to claim 1, wherein the variable that is  
5 controlled is the superficial velocity of the gas in the fluidized bed.

27. A process according to claim 26, wherein the model for the  
control of the superficial velocity of the gas in the fluidized bed uses the  
following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- 10 · Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of the recycle stream;
- Flow rate of inert diluent(s);
- 15 · Opening of the purge valve.

28. A process for the controlled production of polyethylene  
and its copolymers in gas phase reactor(s), isolated or combined, under  
polymerization conditions, in the presence of hydrogen, oxygen, inert  
diluent(s) and a Ziegler-Natta catalyst, wherein said process comprises the  
20 steps of:

a) In the laboratory, periodically testing the  $MFR_2$  and density  
of the resin;

b) Establishing desired values or limits for the set of CVs and  
CCVs which comprise production rate, the resin  $MFR_2$  and density, catalyst  
25 productivity, opening of the temperature control valve, monomer partial  
pressure, superficial velocity of the gas in the reactor, bubble point of the

recycle stream and difference between the inlet and outlet temperature of the water in the cooling system of the reactor;

c) Determining in real time the value of the variables of item (b), the resin properties having their value continuously inferred with the aid of mathematical models;

d) Establishing limits for the set of MVs which comprises flow rate of catalyst, ratio between the concentrations of one or more comonomers and ethylene, ratio between the flow rates of one or more comonomers and ethylene, temperature of the fluidized bed, ratio between the flow rates of oxygen and ethylene, ratio between the concentrations of hydrogen and ethylene, ratio between the flow rates of hydrogen and ethylene, flow rate of the recycle stream, flow rate of inert diluent(s) and opening of the purge valve; reactor pressure, level of the fluidized bed, density of the gas in the recycle stream and superficial velocity;

e) Establishing limits for the variation rate of the variables cited in item (d);

f) By using mathematical models of the process, simultaneously calculating the values which should be taken in a stationary condition by the variables cited in item (d), considering that the level of the fluidized bed and the reactor pressure will remain constant, so that the desired values established in (b) will be reached without violating the limits established in items (b) and (d);

g) Determining the sequence of adjustments to be made along a predetermined period of time, so that the variables cited in item (s) reach the desired values calculated in item (f) considering the dynamics and the relevance of each of the variables cited in (b) and the constraints

established in (e);

h) Adjusting the flow rates of catalyst, comonomer(s), cooling water, oxygen, hydrogen, inert diluent(s), the flow rate of the recycle stream and the opening of the purge valve so as to satisfy the previous  
5 items;

i) based on the laboratory tests, periodically correcting the value inferred for the resin properties.

29. A process according to claim 28, wherein the gas phase reactors are vertical or horizontal fluidized bed reactors or vertical or  
10 horizontal stirred-bed reactors.

30. A process according to claim 28, wherein the mathematical models may be empirical, rigorous or a combination of empirical and rigorous models.

31. A process according to claim 28, wherein the empirical  
15 mathematical models are neural networks.

32. A process according to claim 28, wherein the ethylene is polymerized alone or combined with one or more olefin monomers.

33. A process according to claim 32, wherein the olefin comonomers are propene; 1-butene; 2-methyl propene; 1-pentene; 1-  
20 hexene; 1-heptene; 1-octene; 1-decene; 3-methyl 1-butene; 4-methyl 1-pentene; cyclopentene.

34. A process according to claim 28, wherein the inert diluents are nitrogen; argon and alkanes having up to 6 carbon atoms, isolated or admixed with each other in any amount.

25 35. A process according to claim 14, wherein the variable that is controlled is the production rate.

36. A process according to claim 35, wherein the model for the control of the production rate uses the following input variables:

- Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- 5     · Temperature of the fluidized bed;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- 10    · Level of the fluidized bed.

37. A process according to claim 28, wherein the physical property of the resin that is controlled is the melt flow rate.

38. A process according to claim 37, wherein the model for the control of the melt flow rate uses the following input variables:

- 15     · Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- Ratio between the flow rates of comonomer(s) and ethylene.

39. A process according to claim 1, wherein the physical  
20   property of the resin that is controlled is the density.

40. A process according to claim 39, wherein the model for the control of the density of the resin uses the following input variables:

- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- 25     · Ratio between the flow rates of comonomer(s) and ethylene.

41. A process according to claim 1, wherein the variable that is controlled is the opening of the temperature control valve.

42. A process according to claim 41, wherein the model for the control of the opening of the temperature control valve uses the following  
5 input variables:

- Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of hydrogen and ethylene;
- 10 · Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

43. A process according to claim 1, wherein the variable that is  
15 controlled is the superficial velocity of the gas in the fluidized bed.

44. A process according to claim 43, wherein the model for the control of the superficial velocity of the gas in the fluidized bed uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- 20 · Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- Flow rate of the recycle stream;
- Flow rate of inert diluent(s);
- 25 · Opening of the purge valve.



45. A process according to claim 1, wherein the variables that is controlled is the partial pressure of ethylene in the reactor.

46. A process according to claim 45, wherein the model for the control of the partial pressure of ethylene in the reactor uses the following  
5 input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- 10 · Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

47. A process according to claim 28, wherein the variable that  
15 is controlled is the bubble point of the recycle stream.

48. A process according to claim 47, wherein the model for the control of the bubble point of the recycle stream uses the following input variable:

- Ratio between the flow rates of comonomer(s) and ethylene.

49. A process according to claim 28, wherein the variable that  
20 is controlled is the difference between the inlet and exit temperatures of the water in the cooling system of the reactor.

50. A process according to claim 49, wherein the model for the control of the difference between the inlet and exit temperatures of the  
25 water in the cooling system of the reactor uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- 5   · Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

10       51. A process according to claim 28, wherein the variable that is controlled is the ratio between the concentrations of hydrogen and ethylene in the reactor.

52. A process according to claim 51, wherein the model for the control of the ratio between the concentrations of hydrogen and ethylene in the reactor uses the following input variables:

- 15       · Ratio between the flow rates of comonomer(s) and ethylene;
- Ratio between the flow rates of oxygen and ethylene;
  - Ratio between the flow rates of hydrogen and ethylene;
  - Flow rate of inert diluent(s);
  - Opening of the purge valve.

20       53. A process according to claim 28, wherein the variable that is controlled is the ratio between the concentrations of comonomer(s) and ethylene in the reactor.

54. A process according to claim 53, wherein the model for the control of the ratio between the concentrations of comonomer(s) and  
25   ethylene in the reactor uses the following input variables:

- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of oxygen and ethylene;
- Ratio between the flow rates of hydrogen and ethylene;
- 5   · Flow rate of inert diluent(s);
- Opening of the purge valve.

55. A process according to claim 28, wherein the variable that is controlled is the catalyst productivity.

56. A process according to claim 18, wherein the model for the  
10   control of the catalyst productivity uses the following input variables:

- Flow rate of catalyst;
- Ratio between the flow rates of comonomer(s) and ethylene;
- Temperature of the fluidized bed;
- Ratio between the flow rates of hydrogen and ethylene;
- 15   · Flow rate of inert diluent(s);
- Reactor pressure;
- Opening of the purge valve;
- Level of the fluidized bed.

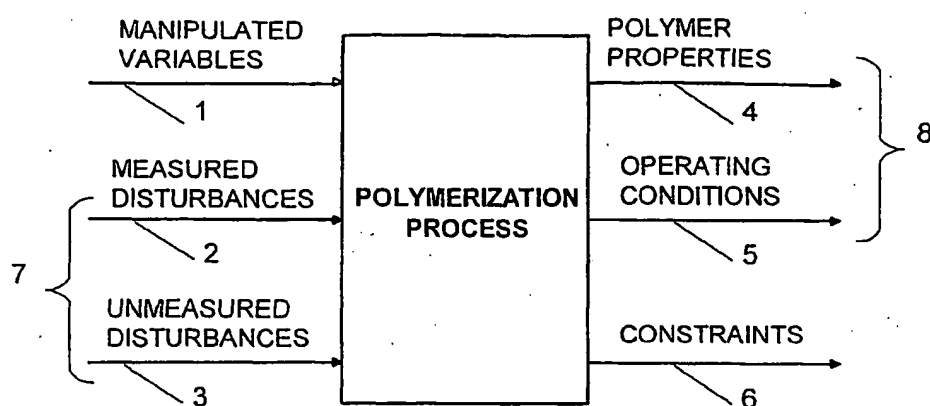


FIGURE 1

		CONTROLLED VARIABLES			CONSTRAINTS	
		CV1	CV2	CV3	CCV1	CCV2
MANIPULATED VARIABLES	MV1	X		X	X	
	MV2		X	X		X
	MV3	X	X		X	
	MV4	X		X		
DISTURBANCES	DV1		X		X	
	DV2	X				X
	DV3				X	

FIGURE 2

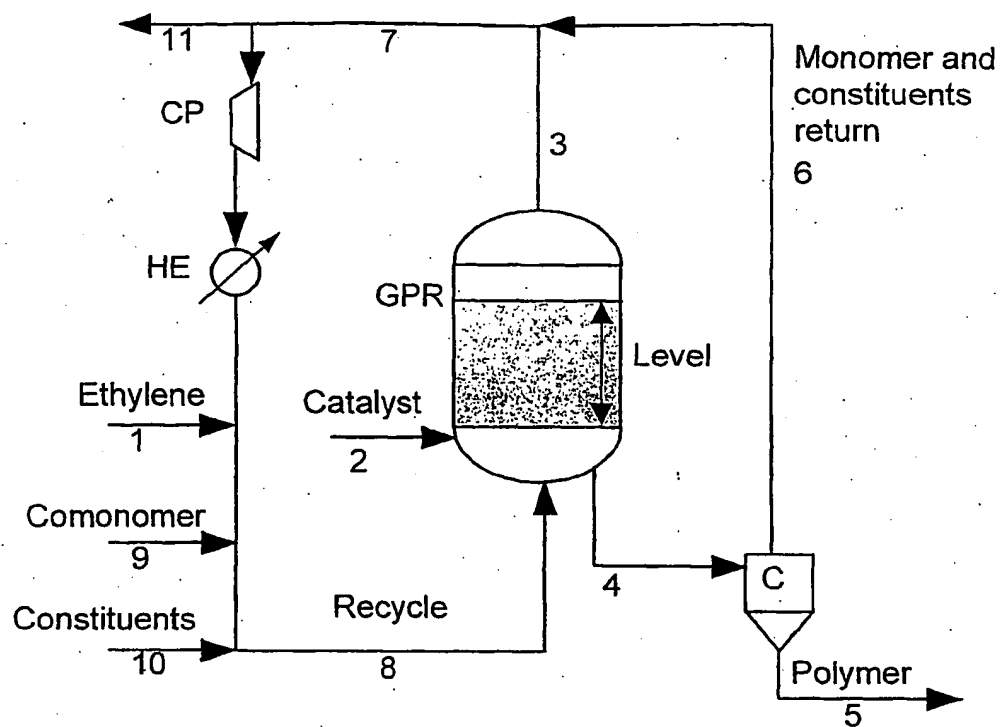


FIGURE 3

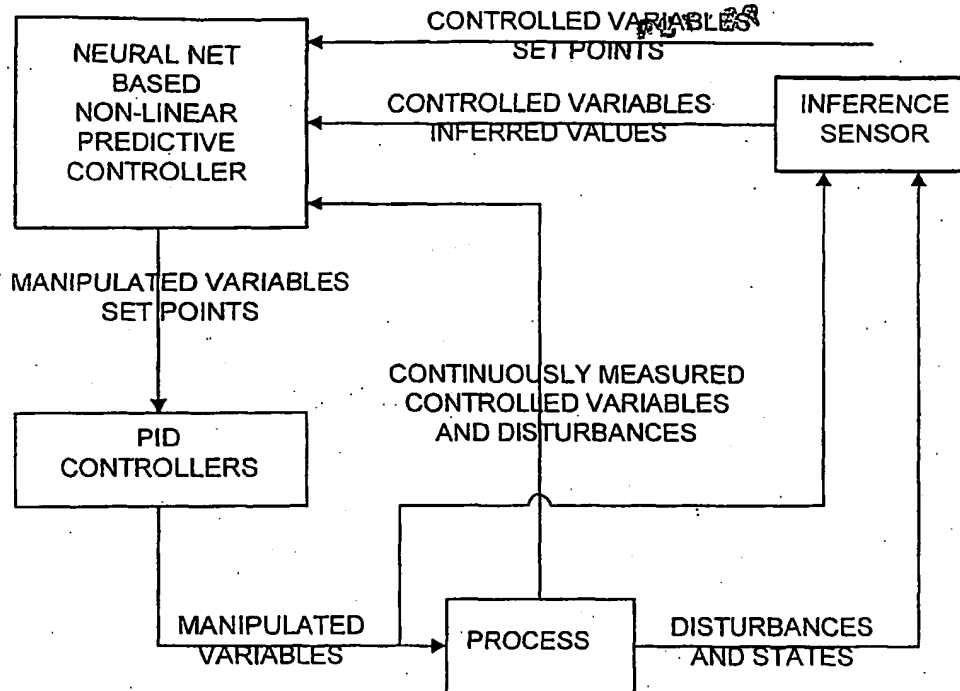


FIGURE 4

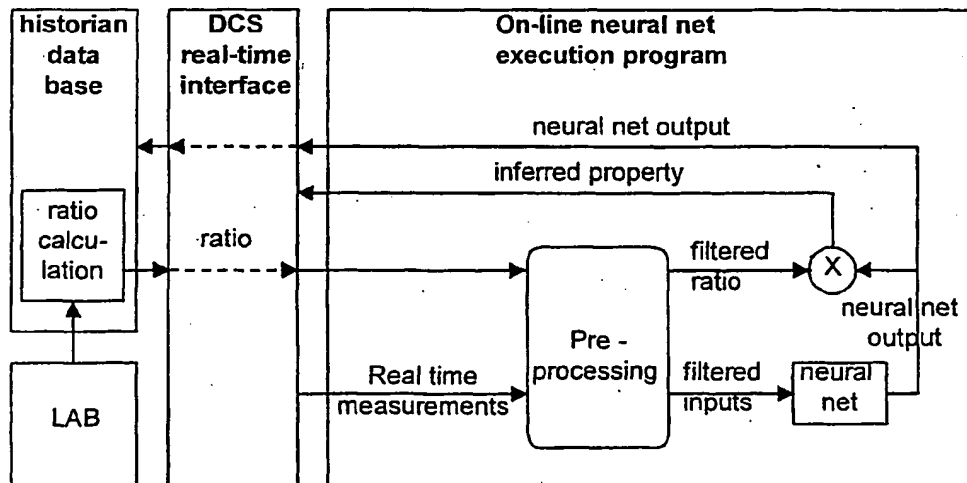


FIGURE 5

		controlled variables / constraints									
		PRODUCTION	MFR <sub>1</sub>	MFR	DENSITY	PRODUCTIVITY	TV	PP	[H <sub>2</sub> ]/[C <sub>2</sub> =]	[C <sub>x</sub> ]/[C <sub>2</sub> =]	SUP VEL
manipulated variables/disturbances	CAT	X				X	X				
	C <sub>x</sub> =/C <sub>2</sub> =	X				X	X	X	X	X	X
	TEMP	X	X	X	X	X	X	X		X	X
	RO		X	X	X			X	X	X	X
	H <sub>2</sub> /C <sub>2</sub> =	X				X	X	X	X	X	X
	RECYCLE										X
	PRESSURE	X				X	X	X			
	BO	X				X	X	X	X	X	X
	IN	X				X	X	X	X	X	X
	LEVEL	X				X	X				
	[H <sub>2</sub> ]/[C <sub>2</sub> =]		X	X							
	[C <sub>x</sub> ]/[C <sub>2</sub> =]		X	X	X						

FIGURE 6

		controlled variables / constraints										
		PRODUCTION	MFR <sub>2</sub>	DENSITY	TV	SUP VEL	PP	BP	DELTA T	[H <sub>2</sub> ]/[C <sub>2</sub> =]	[C <sub>x</sub> =]/[C <sub>2</sub> =]	PRODUCTIVITY
manipulated variables/disturbances	RECYCLE					X						
	PRESSURE	X			X		X		X			
	TEMP	X	X	X	X		X		X	X		X
	LEVEL	X			X		X		X	X	X	
	PP									X	X	X
	CAT	X			X		X		X	X	X	
	IN	X			X		X		X	X	X	
	C <sub>x</sub> =/C <sub>2</sub> =	X			X		X	X	X	X	X	
	H <sub>2</sub> /C <sub>2</sub> =	X			X		X		X	X	X	
	BO	X			X		X		X	X	X	
	[C <sub>x</sub> =]/[C <sub>2</sub> =]		X	X								X
	[H <sub>2</sub> ]/[C <sub>2</sub> =]		X	X								
	GAS DENS					X						

FIGURE 7



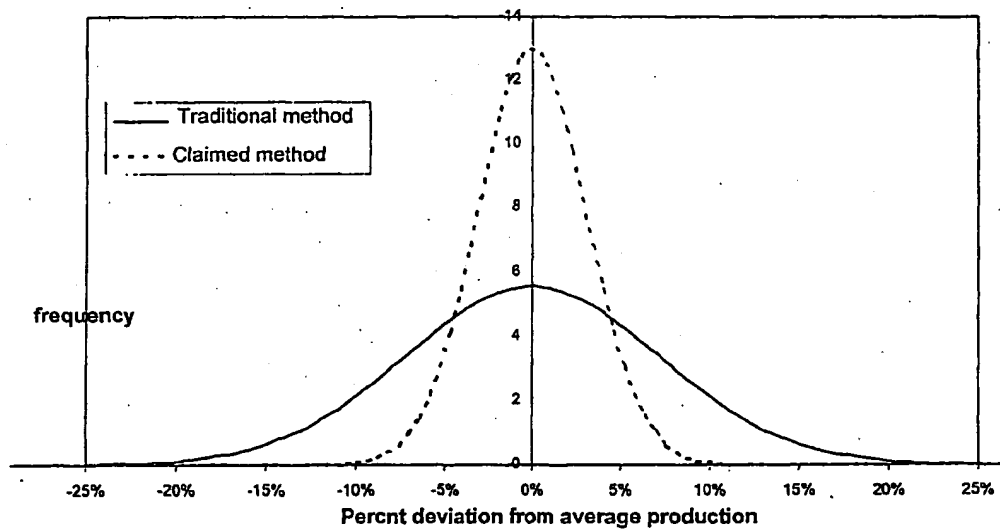
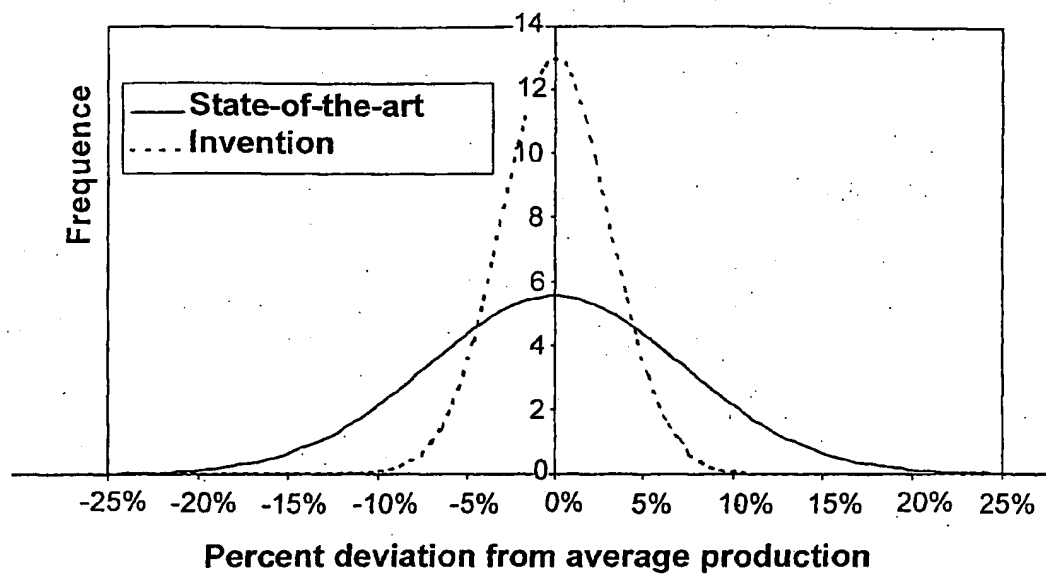


FIGURE 8

# INTERNATIONAL SEARCH REPORT

Int. nat. Application No.  
PCT/BR 00/00155

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 C08F10/02 C08F2/00 605D21/02 G05B13/02		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC 7 C08F		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data, PAJ		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 54231 A (UNION CARBIDE CHEM PLASTIC) 3 December 1998 (1998-12-03)	1-56
Y	page 6, line 6 -page 13, line 15; claims; figures	1-56
X	US 5 844 054 A (HARTLEY IVAN JEREMY ET AL) 1 December 1998 (1998-12-01) cited in the application column 4, line 61 -column 8, line 51; claims	1-27
Y	US 4 469 853 A (MORI ADO) 4 September 1984 (1984-09-04) cited in the application column 3, line 63 -column 4, line 68 column 6, line 5 - line 20; claims	1-56
-/--		
<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
* Special categories of cited documents: <div style="display: flex; justify-content: space-between;"> <div style="width: 48%;"> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 48%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"8" document member of the same patent family</p> </div> </div>		
Date of the actual completion of the international search  <div style="text-align: center; font-weight: bold;">20 March 2001</div>		Date of mailing of the international search report  <div style="text-align: center; font-weight: bold;">05/04/2001</div>
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel (+31-70) 340-2040, Tx 31 651 epo nl Fax (+31-70) 340-3016		Authorized officer  <div style="text-align: center; font-weight: bold;">Kaumann, E</div>

# INTERNATIONAL SEARCH REPORT

In Application No  
PCT/BR 00/00155

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 93 24533 A (AMOCO CORP) 9 December 1993 (1993-12-09) page 6, line 33 -page 8, line 21 page 13, line 28 -page 15, line 23; claims	1-56
A	WO 99 53387 A (ASPEN TECHNOLOGY INC) 21 October 1999 (1999-10-21) claims	1-56

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/BR 00/00155

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9854231 A	03-12-1998	US 5990250 A	23-11-1999
		AU 722190 B	27-07-2000
		AU 7803198 A	30-12-1998
		BR 9809707 A	11-07-2000
		EP 0986587 A	22-03-2000
US 5844054 A	01-12-1998	AU 721083 B	22-06-2000
		AU 5840598 A	24-09-1998
		BR 9800881 A	14-12-1999
		CA 2232207 A	21-09-1998
		EP 0866077 A	23-09-1998
		JP 10265513 A	06-10-1998
US 4469853 A	04-09-1984	JP 55142008 A	06-11-1980
		AT 28879 T	15-08-1987
		DE 3072005 D	17-09-1987
		EP 0027474 A	29-04-1981
		WO 8002288 A	30-10-1980
WO 9324533 A	09-12-1993	CA 2136224 C	24-11-1998
		CN 1081683 A	09-02-1994
		DE 69308209 D	27-03-1997
		DE 69308209 T	12-06-1997
		EP 0642534 A	15-03-1995
		ES 2097520 T	01-04-1997
		HU 70863 A	28-11-1995
		JP 2675919 B	12-11-1997
		JP 7504708 T	25-05-1995
		KR 148257 B	17-08-1998
		RU 2122005 C	20-11-1998
		TR 27889 A	11-10-1995
		US 5504166 A	02-04-1996
WO 9953387 A	21-10-1999	US 6093211 A	25-07-2000
		EP 1070281 A	24-01-2001